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# A Comparison of the Effects of Ageforming on the Precipitation Behaviour in 2xxx, 6xxx and 7xxx Aerospace Alloys

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## Abstract

The aerospace industry wishes to reduce manufacturing costs by the wider application of creep/age-forming. For damage tolerant applications, this requires the introduction of alternative alloys that can be used in artificially aged tempers. An important consideration, in this context, is the effect of the dynamic ageing conditions on the microstructural development of the alloy being formed. The effect of stress level has been investigated by TEM during ageing two commercial aluminium alloys, 6056 and 7475, and one new experimental 2xxx series alloy with good age-formability, to a damage tolerant temper. It has been found that the 2xxx alloy was very sensitive to the age-forming process, whereas in 6056 and 7475 there is little effect.

## 1. Introduction

Age-forming is a relatively novel process that involves simultaneous forming and artificial ageing. In age-forming the alloy sheet (or plate) is deformed, mainly elastically, onto a former by mechanical clamping, or vacuum bagging, in an autoclave and then artificially aged. Stress relaxation occurs by creep, producing the required permanent deformation of the component [1]. In certain alloy systems, it is known that the application of a stress during ageing can significantly alter the material's microstructure [2-6]. An important effect is the preferential nucleation, and possibly growth, of precipitates on specific habit plane variants aligned with the far field stress, which can result in anisotropic mechanical properties [6-7]. The precipitating phases present in different alloy systems have diverse morphologies, habit plane relationships, and positive or negative misfits with the matrix, and therefore maybe expected to react differently during the age-forming process. To date, published data has focused primarily on phases formed in Al-Cu alloys [2-6]. Studies performed on binary Al-Cu and Al-Cu-Mg-Ag alloys have shown that after stress ageing both $\theta''/\theta'$ and $\Omega$ plates can be preferentially orientated by an externally applied stress. Simulations have also shown that the preferred orientation is primarily caused during nucleation [5]. It has further been reported that there is a temperature and composition dependent threshold stress that needs to be exceeded for preferential nucleation to occur on a particular habit plane variant [3-4].

As far as the authors are aware, little has yet been published on the influence of stress ageing on precipitation in 6xxx, 7xxx and 2xxx alloys, which are of interest for commercial aerospace applications of the age-forming technique. The objective of the present study was to investigate the effect on the microstructure of an externally applied stress, when ageing relevant examples of these alloy classes to a damage tolerant temper. The materials investigated were: 6056, 7475, and a developmental, Al-Cu-Mg alloy, 2XU with good age-formability [8].

## 2. Experimental Procedure

The nominal compositions of the three alloys investigated in this project are presented in table 1. 2XU and 7475 were supplied as commercially produced 40 and 60 mm thick plate and 6056 as 5mm sheet, in a solution treated and stretched condition (~ 2-3%). Tensile specimens were machined from the ¼ and ¾ thickness position of the thick plate, and from the full sheet thickness for 6056, with a gauge length of 25mm parallel to the rolling direction. The 0.2% proof stress was determined at the aging temperature using a strain rate of $10^{-5}$ $s^{-1}$. Constant stress-ageing treatments were performed in tension at a range of stress levels, using the damage tolerant tempers defined in table 2, simultaneously with stress-free control samples of identical thickness positioned at the same height in the oven. A Philips CM200 TEM, was used to characterise the precipitates formed in the aged specimens. The loading directions for the stress-aged samples were marked on the specimen foils before polishing. The second phase particles in the 7475 alloy were also characterised by SAXS experiments, using the x-ray synchrotron source at the CCLRC Daresbury Laboratory (beam line 2.2).

Table 1. Chemical Compositions of the alloys investigated in weight percent

| Alloy | Cu | Mg | Mn | Zn | Si | Cr | Fe | Al |
|---|---|---|---|---|---|---|---|---|
| **2XU** | 4.6-5.3 | 0.10-0.50 | 0.15-0.45 | 0.20 | <10 | <0.10 | <0.15 | Bal |
| **AA6056** | 0.5-1.1 | 0.6-1.2 | 0.4-1.0 | - | 0.7-1.3 | - | 0-0.08 | Bal |
| **AA7475** | 1.2-1.9 | 1.9-2.6 | - | 5.2-6.2 | 0-0.1 | 0.18-0.25 | 0-0.12 | Bal |

Table 2. Damage tolerant tempers and ageing heat treatments used for each alloy. A ramp of 75°C/hr was used in all cases.

| Alloy | Temper | Heat treatment |
|---|---|---|
| **2XU** | T851 | 24 hrs@ 173 °C [9] |
| **AA6056** | T7851 | 6 hrs @175 °C + 13 hrs @190 °C |
| **AA7475** | T7351 | 4 hrs @108 °C + 24 hrs @160 °C |

## 3. Results and Discussion

3.1 2XU (Al-Cu-Mg) Alloy: Conventional Ageing.

The precipitation behaviour in the developmental alloy 2XU has not previously been widely reported. For Al-Cu-Mg alloys in a T8 condition, the main strengthening phases are $\theta'$ ($Al_2Cu$) and the S ($Al_2CuMg$). In 2XU the dominant phase should be $\theta'$ because of the high Cu to Mg ratio [10]. The ageing behaviour will therefore be expected to mainly follow the same classical sequence seen in a binary Al-Cu alloy (e.g. GP zones $\rightarrow \theta'' \rightarrow \theta' \rightarrow \theta(Al_2Cu)$). Due to the presence of the Mg and Si, there is also the potential for the formation of other minor phases, such as S ($Al_2CuMg$), $\Omega$ ($Al_2Cu$), and $\sigma$ ($Al_5Cu_6Mg_2$) [11]. (Note; S and S′ cannot be readily distinguished due to their similar crystal structures and will be referred to here simply as S [12]).

Under static ageing conditions, when no applied stress or residual stresses are present, a random distribution of precipitates might be expected to form on all habit plane variants. In the T851 temper this was indeed found to be the case, but not when the alloy was under-aged. In Figure 1 TEM images and electron diffraction data are shown from the fully aged alloy (T851) and, as expected, the dominant phase present is θ′. It can be seen in the image in Figure1a, and the SAED pattern in Figure 1e, that all three {001} cube habit plane variants are present in approximately equal densities. Due to the pre-stretch, θ′ was found to nucleate primarily on dislocations. Narrow PFZs were found at the grain boundaries (GB) due to the precipitation of GB θ ($Al_2Cu$).

Although not so easily visible in this orientation, a significant density of Ω ($Al_2Cu$) plates was also observed, highlighted in Figure 1c. Four quite strong Ω diffraction spots can be seen symmetrically arranged around the $\{110\}_\alpha$ position (at 1/3 and 2/3 g{220}) in the magnified insert in Figure 1e.

The similar intensities of these reflections indicate that Ω is also randomly distributed over the four possible {111} habit plane variants. Low densities of S ($Al_2CuMg$) and σ ($Al_5Cu_6Mg_2$) were also identified. A small number of lath shaped S precipitates can be seen in the magnified dark-field images in Figure1b and a cubic σ ($Al_5Cu_6Mg_2$) particle is shown in Figure 1d.

Even when aged without an externally applied stress, with short aging times the GPZs and θ″ plates in the 2XU alloy were found to be preferentially aligned with respect to the rolling direction, as can be seen in Figure 2. However, this alignment was found to disappear when small samples of the alloy were re-solutionised, and re-aged. This effect can therefore be attributed to the presence of weak residual stresses in the thick commercial plate and the high sensitivity of nucleation on preferred habit plane variants by GPZs/θ″ in the presence of a far field stress [3]. This behaviour is discussed further below for the stress-aged samples.

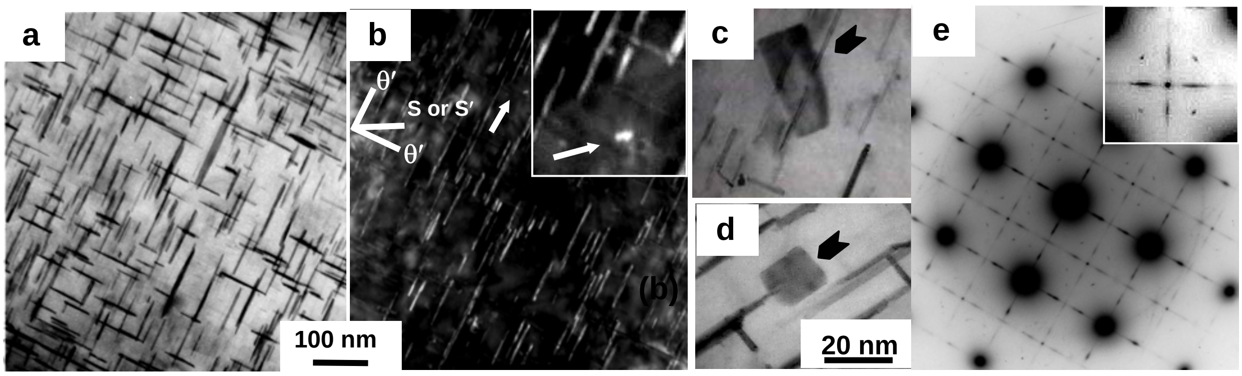


Figure 1: TEM micrographs of 2XU after aging to T851 without an applied stress, (a) mainly θ′ precipitates, (b) DF image showing one variant of the θ′ and a few S ($Al_2CuMg$) precipitates (magnified), (c) an example Ω ($Al_2Cu$) plate, (d) σ ($Al_5Cu_6Mg_2$), and (e) a SAED pattern indicating no preferential alignment of θ′, or S. The magnified insert shows four symmetric reflections around the $\{110\}_\alpha$ positions from Ω, again indicating no preferred alignment. The electron beam is near a $<001>_\alpha$ zone axis.

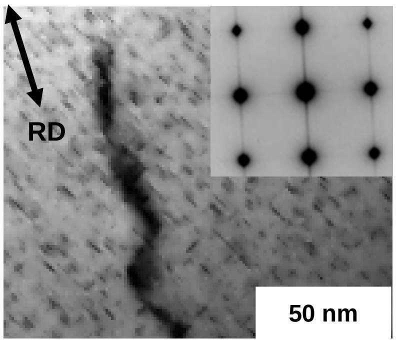


Figure 2 GPZs/θ″in the 2XU alloy after 10 minutes at temperature, following ramp heating at 75°C/hr with no externally applied stress.

### 3.2 2XU (Al-Cu-Mg) Alloy: Stress-Ageing.

After ageing at a range of stress levels from 30% to 90% of yield (67-202 MPa), to the T851 condition, the 2XU alloy was found to contain the same phases observed in the conventionally heat-treated specimen (i.e. θ′, Ω, S and σ). However, at higher stress levels strong alignment of the θ′ plates with the tensile stress axis was observed. This can be seen from the example bright field images shown in Figure 3, orientated near a <001> zone axis, as well as the diffraction pattern inserts magnified from around the $\{110\}_{\alpha}$ position, which show strong preferential streaking along the $[100]_{\alpha}$ direction aligned closest to normal to the stress axis.

Even at the highest stress level (Figure 3d), there was still faint streaking along the $[010]_{\alpha}$ direction nearest to parallel with the tensile axis, indicating that 100% alignment did not occur. It should be noted that only two plate variants can be seen in this orientation. The plate lying in the plane of the page produces a strong reflection at the $\{110\}_{\alpha}$ position at high stress levels, as this plate variant is also aligned preferentially with the tensile stress axis. The θ′ plates were further found to have a higher density and smaller size, on the favoured habit plane variants closet to being parallel with the stress axis, as the stress level increased. Although the stress axis is rarely perfectly aligned with a <001> direction in a polycrystalline alloy, after examining several grains in each sample, it was found that above an applied stress level of ~ 150 MPa (Figure 3c &d) the degree of alignment appeared to saturate, and decreased rapidly to become more isotropic at stresses below ~ 50 MPa, in agreement with measurements by Starke and co-authors who found a threshold stress for the alignment of θ′ of ~ 20 MPa [3].

From close inspection of the four Ω reflections on the diagonals around the central $\{110\}_{\alpha}$ position, in the diffraction patterns shown in Figure 3, it can be seen that their distribution becomes asymmetric as the stress level increases above ~ 112 MPa, with the progressive strengthening and weakening of the pairs of reflections on opposing diagonals. This indicates that Ω precipitation also occurs preferentially on the two habit plane variants closest to being parallel with the tensile stress axis. On ageing with an applied stress of 112 MPa and below, no preferred alignment could be demonstrated for Ω and the threshold stress for alignment appears to be much greater than for θ′, in agreement with the observations of Skrotzki et al. [3] who found it to occur between120-140 MPa. The crystal structure of the Ω phase is still controversial, but is probably closely related to the θ phase which nucleates semicoherently in the presences of trace elements that relax the interfacial misfit [13]. Both θ′ and Ω have a ‘c’ axis that shows poor fit with the α matrix and consequently nucleate and grow as thin plates to minimise the misfit strain. The smallest misfit of the θ' phase is achieved with a nucleus two unit cells thick, which gives a vacancy-type misfit of – 4.5%. Although the unit cell of Ω is less well known, the misfit is again negative and probably larger than that for θ′ [3]. Any misfitting coherent or semi-coherent precipitate will attempt to nucleate and grow in such a way as to minimise its strain energy. It is now well established (e.g. [3-4]) that, because of the negative misfit, θ′ precipitates 'prefer' to nucleate along the cube planes experiencing a compressive strain.

The effect of an applied tensile stress is therefore to reduce the vacancy misfit for the θ′ precipitate plates when they nucleate on planes parallel to the stress axis, which on average in a polycrystalline alloy biases precipitation on two of the possible three $\{100\}_\alpha$ planes. A similar behaviour is observed for the Ω phase which forms preferentially on the two habit plane variants most closely aligned with the tensile stress axis, of the four

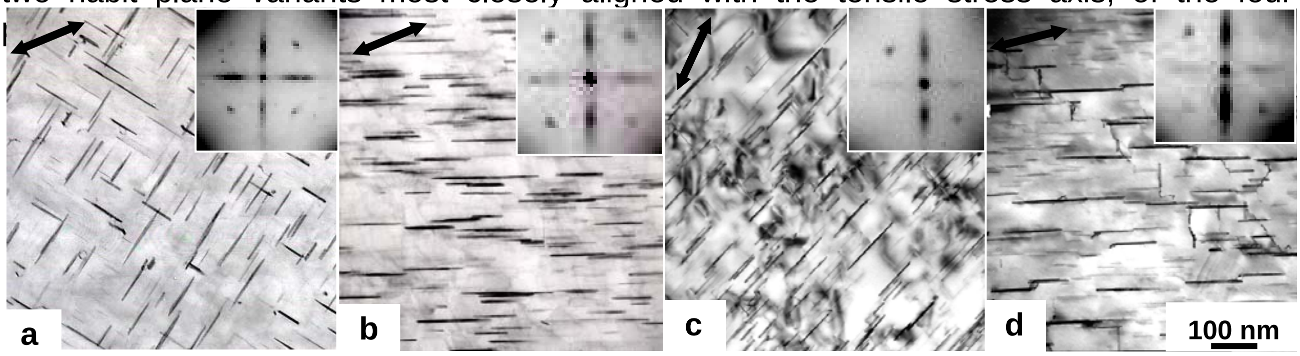


Figure 3: TEM micrographs and the magnified region around the {110} position, taken from SAED patterns (inserts), for the 2XU alloy after stress-ageing to T851 for increasing stress levels of: (a) 67 MPa (30%$\sigma_y$), (b) 112 MPa (50%$\sigma_y$), (c) 157 MPa (70%$\sigma_y$), and (d) 202 MPa (90%$\sigma_y$). The electron beam is near a $<001>_\alpha$ zone axis. The arrows indicate the projected tensile axis. Note the SAED patterns are rotated relative to the image for ease of presentation.

A number of previous studies and modelling simulations have shown that for θ″ and θ′ only a small deviatoric perturbation in the far field stress is sufficient to increase the nucleation rate for a habit plane, sympathetically aligned with respect to the stress axis, relative to the other possible variants [2-7]. In comparison, it is predicted that much higher stresses are required to bias growth [5]. Although, in this case, the situation is complicated by the misfit of the structural growth unit during ledge migration, which depends on its height.

The results for the conventionally aged sample indicate that small residual stresses within thick plate materials can be sufficient to bias the nucleation of GPZs/θ″, but not θ′. This suggests θ′ has a higher stress threshold. However, GPZs nucleate homogeneously, and in the stretched alloy studied, θ' nucleated predominately within the strain field of dislocations, which might be expected to reduce the sensitivity of nucleation to an external stress. Although more work is required to confirm this, the threshold for alignment in the current study also appeared higher than in previous work on unstretched samples [3,4].

The fact that Ω has a higher misfit than θ′, might suggest that it should have a lower stress threshold than observed. However, nucleation of Ω is complex and appears to involve the formation of a monolayer of solute (Mg and/or Ag) at the interface, which reduces the misfit [13], and this may decrease its sensitivity to the external stress.

In 2XU there is also the possibility that the minor S and σ phases can become aligned due to the applied stress field. Although, the symmetric cubic morphology of σ ($Al_5Cu_6Mg_2$)

phase makes stress alignment appear unlikely. No obvious alignment of S or σ was observed, but their low density and the large number of habit plane variants for S made it very difficult to determine if any systematic alignment had occurred.

## 3.3 AA6056 Conventional and Stress-Ageing.

The aging behaviour of quaternary Al-Mg-Si-Cu alloys, such as 6056 and 6013, is generally thought to involve two simultaneous precipitation sequences; *$SSSS_\alpha \rightarrow$GP zones$\rightarrow\beta''\rightarrow\beta'\rightarrow\beta$ ($Mg_2Si$)* and *$SSSS_\alpha\rightarrow$ GP zones$\rightarrow$ $\lambda'$ (or B′, Q′)$\rightarrow$ $\lambda$ (or B, Q) ($Al_5Cu_2Mg_8Si_6$)* [14-15]. TEM investigation of this alloy, in the T7851 temper, showed that it predominantly contained β′ rods, and λ′ as laths and some globular morphology precipitates (Figure 4), in agreement with results reported by Tanaka and Warner [14]. No preferential alignment, or any other difference in these phases was observed, in the stress-aged 6056 specimens, even when the maximum possible stress was applied without the sample failing (199.5 MPa). When comparing the BF images in Figure 4a & b, there is no indication of any single precipitate variant dominating in one direction.

Equally, close examination of the streaks from the β′ (black arrows) and λ′ precipitates (white arrows) in the <001> zone SAD patterns shows the same symmetrical distribution of intensities along the [100] and [010] directions. It can therefore be concluded that an externally applied tensile stress has little effect on the ageing characteristics of the 6056 alloy.

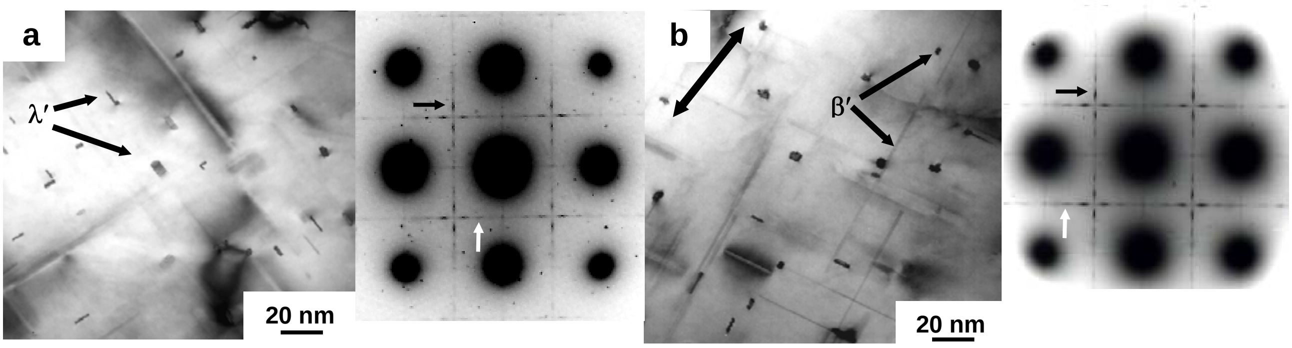


Figure 4: TEM micrographs and SAED patterns from 6056 aged to T7851 showing β′ and Q′ precipitates, (a) conventionally and (b) stress-aged under a constant 199.5 MPa. The electron beam is near a $<001>_\alpha$ zone axis. The double-headed arrows indicate the projected load direction.

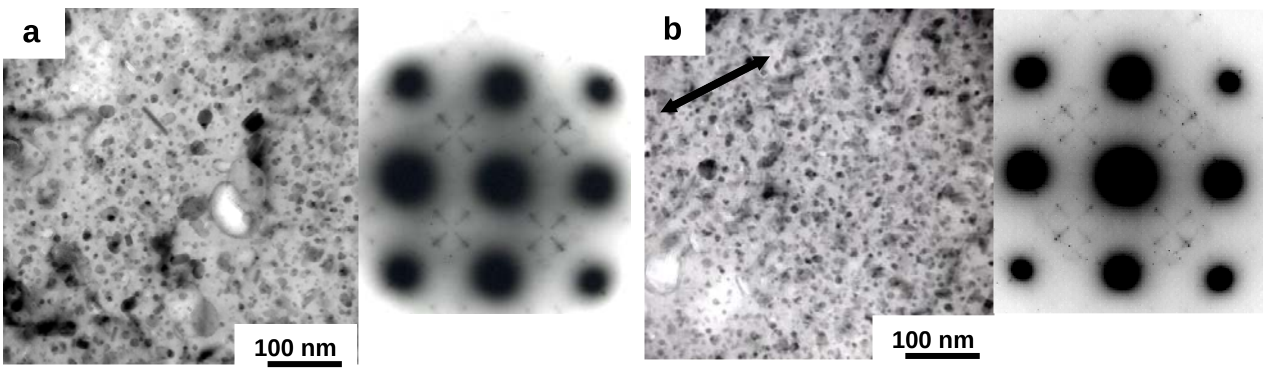


Figure 5: TEM micrographs and $<001>_\alpha$ SAED patterns of 7475 aged to T7351 (a) conventionally and (b) stress-aged under a constant 245 MPa tensile stress

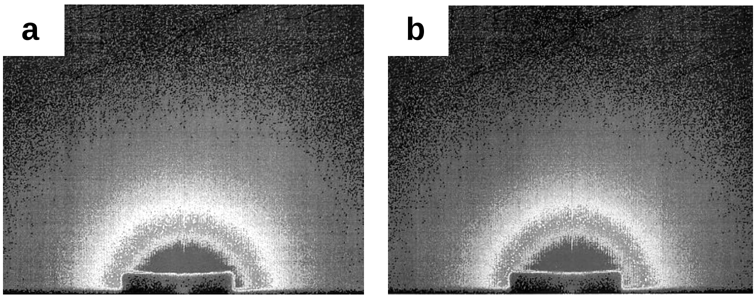


Figure 6: SAXS diffraction patterns from 7475 aged to T7351 (a) conventionally and (b) stress-aged under a constant 245 MPa tensile stress.

3.4 7475 Conventional and Stress-Ageing.

The precipitation sequence in high strength Al-Zn-Mg-Cu alloy is well known as; $SSSS_{\alpha} \rightarrow$GP zones$\rightarrow\eta'\rightarrow\eta$ ($MgZn_2$) [16,17]. After ageing to the T7351 temper the 7475 alloy contained a fine distribution of $\eta'$, that develops from GPZs that grow during the first aging stage, and equilibrium $\eta$, nucleated at dislocations and dispersoids, as can be seen from Figure 5.

Similar to 6056, the TEM data showed no obvious difference in precipitate density, size or alignment between the conventional and stress aged samples, even when the maximum possible stress was applied without the sample failing 245 MPa. In the BF images in Figure 5 there is no indication of any single precipitate variant dominating in any particular direction. The intensities of the $\eta$ reflections along the two $[110]_{\alpha}$ directions present in the [001] zone axis are also symmetrically distributed and have the same intensity in each sample. To confirm the TEM results, SAXS measurements were also performed (Figure 6). This data has not yet been full analysed, but simple comparison of the conventional and stress-aged materials shows that both patterns are very similar and give close to isotropic intensities, indicating that there is no strong preferred alignment present. Using a spherical model to fit the intensities gave an average particle size of 4.8 and 5.0 nm for the conventionally and stress aged samples, which is within the experimental error.

## 4. Conclusions

The effect on the precipitation behaviour of an externally applied stress during ageing has been compared for a typical alloy from each class under investigation for age-forming aerospace components. It was found that only the thin plate shaped precipitates, $\theta''$, $\theta'$ and $\Omega$, present in Cu rich 2xxx alloys, were affected by the far field stress applied during age-forming. In contrast, no effect was observed for the lath shaped Q' and rod/lathe $\beta'$ found in Al-Mg-Si-Cu alloys, or the approximately rhombohedral morphology $\eta'$ phase in Al-Zn-Mg-Cu alloys. As a consequence, the microstructure of alloys like 6056 and 7475 should be unaffected by the age-forming process.

For the $\theta''$, $\theta'$ and $\Omega$ plate shaped precipitates, the applied stress reduces the plate normal vacancy misfit strain of the nucleus, biasing nucleation on habit plane variants, which experience a compressive strain. In tensile loading this leads to preferential precipitation on two of the possible three habit {100} planes for $\theta''/\theta'$ and two of the four {111) planes for $\Omega$, which are aligned closest to the stress axis. The threshold stress required is very low in the case of θ″/θ′ and considerably higher for Ω. The high sensitivity to the stress is related to the asymmetric strain field of the nucleus, which in the case of θ″ is known to develop from disc shaped Cu GPZs on {100} planes. For the other phases, such as η′ and β', which grow from more spherical morphology GPZs, or solute clusters, there is clearly less potential for the nucleation event to be affected by an externally applied stress.

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